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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

AD-A218 569

DTIC ELECTE 1990

1a. REPORT SECURITY CLASSIFICATION: Unclassified	1b. RESTRICTIVE MARKINGS
2a. SECURITY CLASSIFICATION AUTHORITY	3. DISTRIBUTION/AVAILABILITY OF REPORT: Approved for public release; distribution is unlimited.
	5. MONITORING ORGANIZATION REPORT NUMBER(S): AFOSR-TR- 90-0262
6a. NAME OF PERFORMING ORGANIZATION: Yale University	6b. OFFICE SYMBOL (If applicable)
7a. NAME OF MONITORING ORGANIZATION: AFOSR/NA	
6c. ADDRESS (City, State, and ZIP Code): New Haven CT 06520	7b. ADDRESS (City, State, and ZIP Code): Building 410, Bolling AFB DC 20332-6448
8a. NAME OF FUNDING/SPONSORING ORGANIZATION: AFOSR/NA	8b. OFFICE SYMBOL (If applicable): NA
9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER: AFOSR 88-0100	
8c. ADDRESS (City, State, and ZIP Code): Building 410, Bolling AFB DC 20332-6448	10. SOURCE OF FUNDING NUMBERS

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT ACCESSION NO.
61102F	2308	A3	

11. TITLE (Include Security Classification)
(U) Nonlinear Spectroscpy of Multicomponent Droplets and Two- and Three-Dimensional Measurements in Flames

12. PERSONAL AUTHOR(S)
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13a. TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT (Year, Month, Day)	15. PAGE COUNT
Annual Technical	FROM 1/1/89 TO 12/31/89	1990, January, 15	17

16. SUPPLEMENTARY NOTATION

17. COSATI CODES		
FIELD	GROUP	SUB-GROUP

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
Nonlinear optical diagnostics, Fluorescence imaging, Stimulated Raman scattering (Continued)

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

Our progress in the area of nonlinear spectroscopy of droplets includes the following: (1) development of a fluorescence imaging technique which is capable of demarcating the liquid phase of the deformed and ejected droplets from the vapor phase of the ejected material; (2) determination that the absorption of the laser-induced plasma quenches the stimulated Raman process and sets an upper limit on the incident intensity which can be used to generate stimulated Raman scattering (SRS) within droplets; (3) completion of a statistical study of the SRS from single droplets which are irradiated at a fixed input laser intensity (from a single-mode or a multimode laser); (4) conclusion that, for single-mode laser excitation, SRS is pumped by the stimulated Brillouin radiation, not directly by the laser radiation; and (5) initiation of studies on the phase-matching requirement for four-wave mixing processes in droplets and on the phase velocity of waves on a morphology-dependent resonance. Keywords

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT: ☒ UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT ☒ DTIC USERS	21. ABSTRACT SECURITY CLASSIFICATION: Unclassified
22a. NAME OF RESPONSIBLE INDIVIDUAL: Julian M Tishkoff	22b. TELEPHONE (Include Area Code): (202) 767-0465 — 22c. OFFICE SYMBOL: AFOSR/NA

DD Form 1473, JUN 86 — Previous editions are obsolete.

SECURITY CLASSIFICATION OF THIS PAGE
Unclassified

18. Subject Terms (Continued):

Phase-matching conditions,
Phase velocity of modes,
Laser diagnostics,
Turbulent flames,
Premixed flames,
Three-dimensional flow measurements,
Burning velocity,

19. Abstract (Continued):

Progress in two- and three-dimensional measurements in flames includes new results in the following: (1) measurement of differential diffusion effects in turbulent jets, (2) development of a simple Rayleigh scattering technique for visualizing supersonic flows, and (3) two-dimensional measurements of the time evolution in a turbulent premixed flame, and (4) the development of a new technique for three-dimensional measurement of time development in turbulent flows.

Annual Report

to the

Air Force Office of Scientific Research

NONLINEAR SPECTROSCOPY OF MULTICOMPONENT DROPLETS AND TWO- AND THREE-DIMENSIONAL MEASUREMENTS IN FLAMES

AFOSR Grant No. 88-0100

January 1 - December 31, 1989

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January 1990

Distribution of this report is unlimited.

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NONLINEAR SPECTROSCOPY OF MULTICOMPONENT DROPLETS

RESEARCH OBJECTIVES

Following is a brief description of the three principal research objectives related to nonlinear spectroscopy of liquid droplets, as outlined in our original proposal:

1. To investigate the stimulated Raman scattering (SRS) statistics from single droplets in a flowing linear stream with the following three types of laser excitation: (1) a cw mode-locked Nd:YAG laser which can be pulsed at a rate of 80 MHz, thereby enabling us to average the SRS signal over many laser pulses per second of integration time; (2) a multimode Q-switched Nd:YAG laser which has a linewidth of 0.6 cm^{-1} and numerous picosecond spikes superimposed on the nominal 10 ns Q-switched pulse; and (3) a single-mode Q-switched Nd:YAG laser (with an injection seeder) which has a linewidth of 0.006 cm^{-1} and a temporally smooth pulse of 7-10 ns.

2. To explore the possibility of using CARS spectroscopy to determine the concentration distribution from different regions within the droplet rim. We plan to use a CCD two-dimensional detector to accumulate the CARS signals from numerous laser shots from a single-mode Q-switched laser or a cw mode-locked laser. Particular emphasis will be placed on the nonuniform concentration distribution within a multicomponent fuel droplet which results from combustion, nonuniform heating, and acceleration of the droplet.

3. To determine to what extent the laser pulse deforms the transparent droplet via electrostrictive forces. Knowledge of such laser-induced shape deformation is important in determining the Q factor of the morphology-dependent resonances (MDR's) which provide the necessary optical feedback for SRS within the droplets.

RESEARCH STATUS

Our results for the second year of research on nonlinear spectrsocopy of droplets can be summarized as follows:

1. Laser-Induced Shape Deformation by Laser-Induced Heating and Electrostriction

We began with the last research objective listed above. During the first year, we have demonstrated that shape distortion of totally transparent droplets can result via the electrostrictive force associated with the gradient of the laser intensity (∇I) which is largest at the internal focal spot just within the droplet shadow face. The laser-induced electrostrictive force pushes against the surface tension force of the droplet and causes the droplet to bulge at the shadow face. After several microseconds, the droplet shape oscillates between a spheroid and a sphere, the distortion amplitude finally dampens, and the droplet remains spherical in shape. The shape oscillation is proportional to the square root of the dynamic surface tension of the liquid, and the damping rate is inversely proportional to the bulk viscosity of the liquid. Quantitative information on laser-induced shape distortion is important because this electrostrictive effect sets an upper limit on the amount of laser energy needed to shatter a droplet. Furthermore, knowledge of the electrostrictive effect enables us to estimate the shape distortion amplitude induced by a high intensity laser pulse, which is used to pump the SRS. [Our results were published in Optics Letters 13, 916 (1988)].

During this year, we developed a fluorescence technique which can image the liquid (not the vapor) phase of a droplet and the ejected material after a CO_2 laser pulse nonuniformally heats the droplet. The shapes of the parent droplet and of the ejected liquid material were studied after laser-induced explosive vaporization of water and ethanol droplets, which have large differences in their absorption at 10.6 μm and hence different material ejection characteristics. With this technique, we can visualize the parent droplet and the liquid phase portion of the ejected material without the interfering contribution from the vapor phase portion

of the ejected material. After irradiation of the droplets with a CO_2 laser beam, the droplet shape distortion, ejection, shattering, and propulsion have been photographed using the fluorescence emission from rhodamine 6G dye added to water (1×10^{-4} M) and to ethanol (0.2×10^{-4} M). The fluorescence technique is a variation of an imaging approach initially developed by Prof. Lynn Melton to determine the internal temperature distribution within a droplet using exciplex-monomer fluorescence. [Our results have been submitted to Optics Letters (publication #1) and for the Proceedings of a 1989 AGARD conference (publication #2)].

2. Laser-Induced Breakdown Which Quenches SRS

In keeping with the last research objective, we studied the growth, decay, and quenching of SRS in transparent droplets using a time-resolved spectroscopic technique which consists of a spectrograph placed in front of a framing camera. The spectrograph disperses the radiation from the laser and the first- and ith-order Stokes SRS along the vertical axis, and the framing camera displays the temporal information along the horizontal axis. The time delay between the first-order Stokes SRS and the input laser pulse is the buildup time of the SRS starting from the spontaneous Raman noise within the droplet. The much shorter time delay between the first-order Stokes and the higher-order Stokes is indicative that, for the cascading stimulated Raman process, the initial Raman signal is produced by the four-wave mixing process, not by the spontaneous Raman process as is the case for the first-order Stokes SRS. The various orders of Stokes SRS radiation continue to circulate within the droplet, even after the laser pulse is off. The radiation decay rate is dependent on the leakage rate of the MDR's which provide the optical feedback for the SRS radiation and on the intensity-dependent depletion rate which results from the cascading SRS.

At high input laser intensities, well above the SRS intensity threshold, the laser-induced breakdown of droplets sets another upper intensity limit which should not be exceeded. Laser-induced breakdown is accompanied by the development of a dense, high temperature plasma

which can absorb the SRS. Such breakdown is localized in a region just within the droplet shadow face and is initiated during the rising portion of the laser pulse, which pumps the SRS. Optical absorption of the plasma quenches the SRS which would have been pumped by the subsequent portion of the laser pulse. Consequently, the higher the input laser intensity is, the less SRS is generated once laser-induced breakdown occurs. The onset of laser-induced breakdown is sensitively indicated by the quenching of SRS, which precedes our ability to detect atomic emission from species within the laser-induced plasma. [Our results were published in the Proceedings of the Laser Material and Laser Spectroscopy Conference (publication #3).]

3. Excitation of SRS with Single-Mode and Multimode Q-Switched Lasers

We completed the experimental study of the SRS statistics from single droplets in a flowing stream (our first research objective). We compiled our results in the form of a histogram of the SRS intensity for 700 laser shots at a fixed intensity. The fluctuation of the SRS intensity for single-mode laser pumping was much less than that for multimode laser pumping. Therefore, single-mode laser pumping should always be used to minimize fluctuations in the SRS intensity.

The SRS intensity threshold was also investigated with single-mode and multimode Q-switched Nd:YAG laser excitation. The new findings can be summarized as follows: (1) The SRS intensity threshold with a single-mode laser beam is noted to be three times lower than that with a multimode beam. (2) The intensity threshold for stimulated Brillouin scattering (SBS) from droplets is lower than that for SRS with single-mode excitation. (3) both SBS and SRS appear as equal length arcs which are confined to the droplet illuminated and shadow faces; (4) the SRS and SBS consist of several pulses within the smooth Q-switched laser pulse ($\approx$7 ns duration); (5) the first SBS pulse always occurs sooner than the first SRS pulse; and (6) the temporal profiles of the SRS and SBS pulses, which are simultaneously measured with a streak camera (100 ps resolution), are temporally correlated, i.e., the minimum of the (n + 1)th SBS pulse occurs when the nth SRS pulse reaches a maximum.

Based on the SRS and SBS results, we conclude that the following processes occur within droplets upon irradiation by a single-mode laser beam: (1) the droplet illuminated face enhances the incident intensity in a region just within the droplet shadow face; (2) both spontaneous Brillouin scattering and Raman scattering are created and amplified with optical feedback provided by the droplet morphology; (3) the SBS threshold is exceeded before the SRS threshold; (4) the SBS, rather than the laser radiation, within the droplet serves as the pump for SRS; (5) the first SBS pulse is depleted by pumping the first SRS pulse; (6) the first SRS pulse is depleted by the cascade stimulated Raman process; (7) the second SBS pulse needs to be repumped by the remaining single-mode laser pulse; (8) the second SRS pulse needs to be repumped by the second SBS pulse; and (9) for the third SBS and SRS pulses, the previous sequences are repeated until there is not enough laser intensity to repump the nth SBS pulse. The most striking finding from this phase of our research is that the SRS is pumped by the SBS, not by the laser pulse. [Our findings will be published in the Journal of the Optical Society of America B.]

4. Four-Wave Mixing in Droplets

The second objective of our research is to explore the possibility of using CARS spectroscopy to determine the concentration distribution from different regions within the droplet rim. CARS is one example of the more general four-wave mixing processes which require phase-matching. Numerous phase-matching configuations (such as BOXCARS) have been conceived for plane waves in an extended medium. However, phase-matching is poorly understood tor waves circumnavigating the droplet rim. For example, what is the phase velocity of a MDR, which can be envisioned as two counterpropagating guided waves?

We initiated an experimental and theoretical program to further our understanding of the phase-velocity of guided waves within droplets. We chose to use third-order sum frequency generation (TSFG) as a means of providing more direct information about the phase velocity of MDR's and, more specifically, about the phase-matching ability among waves trapped within the

droplet. The nonlinear source polarization which induces the electric field at the third-order sum frequency is:

$$P^{NLS}(\omega_1 + \omega_2 + \omega_3) = \chi^{(3)} E(\omega_1) E(\omega_2) E(\omega_3),$$

where ω_1, ω_2 and ω_3 are the laser frequency at ω_L and/or the jth-order Stokes SRS frequency at ω_{js}. As in the case for the CARS intensity, the TSFG intensity is proportional to the square of the coherence length $(l_{coh})^2$, where $l_{coh} = \pi/\Delta k$. For the degenerate case (when $\omega_1 = \omega_2 = \omega_3$), Δk for third harmonic generation (THG) is:

$$\Delta k = 3\omega_1\{[v^{MDR}(3\omega_1)]^{-1} - [v^{MDR}(\omega_1)]^{-1}\},$$

where $v^{MDR}(3\omega_1)$ and $v^{MDR}(\omega_1)$ are the phase velocities of MDR's at ω_3 and ω_1, respectively. For the more general case, when $\omega_1 \neq \omega_2 \neq \omega_3$, Δk is only slightly more complicated.

We calculated $v^{MDR}(\omega_1)$ for MDR's of various mode numbers and mode orders and noted that $v^{MDR}(\omega_1) \geq c/n(\omega)$, where c is the speed of light in vacuum and and $n(\omega)$ is the index of refraction of the liquid at ω. The phase matching of MDR's is greatly improved relative to the phase matching of plane waves propagating in the bulk liquid. Our tentative conclusion is that the TSFG intensity can be selectively increased by tuning a particular MDR to $\omega_1 + \omega_2 + \omega_3$. [We are in the process of analyzing our data for submission to Optics Letters and the Journal of the Optical Society.]

PUBLICATIONS

1. A.S. Kwok, C.F. Wood, and R.K. Chang, "Fluorescence Imaging of CO_2 Laser Heated Droplets," submitted to Opt. Lett.

2. Richard K. Chang and Alfred S. Kwok, "High Intensity Laser Beam Interactions with Single Droplets," to be published in the Proceedings of the 1989 AGARD Symposium on Atmospheric Propagation in the UV, Visible, IR and MM-Wave Region and Related Systems Aspects, held in Copenhagen, October 9-13, 1989.

3. J.-B. Zheng, W.-F. Hsieh, S.-C. Chen, and R.K. Chang, "Growth, Decay, and Quenching of Stimulated Raman Scattering in Transparent Liquid Droplets," in Laser Materials and Laser Spectroscopy, Z. Wang and Z. Zhang, eds. (World Scientific, Singapore, 1989), p. 259.

4. J.-Z. Zhang, G. Chen, and R.K. Chang, "Pumping of Stimulated Raman Scattering by Stimulated Brillouin Scattering within a Single Droplet: Input Laser Linewidth Effects," to be published in J. Opt. Soc. Am. B.

PROFESSIONAL PERSONNEL

Alfred Kwok, Graduate Student
Kim Juvan, Graduate Student
Jian-Zhi Zhang, Graduate Student
William Acker, Research Associate

DEGREES AWARDED

None

PATENTS

None

COUPLING ACTIVITIES

Some of the fluorescence imaging, SRS and TSFG results were presented at the following meetings and workshops in 1989:

Fifth Annual Workshop on the Physics of Directed Energy Propagation in the Atmosphere, New Mexico State University, Las Cruces, NM , February 28-March 1.

Conference on Lasers and Electro-Optics (CLEO), Baltimore, MD, April 27 (invited talk).

CRDEC Scientific Conference on Obscuration and Aerosol Research, Aberdeen Proving Ground, MD, June 28-30.

1989 Gordon Research Conference, Plymouth, NH, July 17-21 (served as Chairman).

1989 AGARD Symposium on Atmospheric Propagation in the UV, Visible, IR and MM-Wave Region and Related Systems Aspects, Copenhagen, October 9-13.

TWO- AND THREE-DIMENSIONAL MEASUREMENTS IN FLAMES

RESEARCH OBJECTIVES

Laser diagnostic techniques are being developed that are capable of two- and three-dimensional mapping of scalars in turbulent flames. In addition, we wish to extend our imaging methods to allow the measurement of the temporal evolution of flow structures in two and three dimensions. Whenever possible, the techniques are tailored to measure quantities and flow configurations of current interest to combustion modelers. The availability of quantitative data on the spatial and temporal characteristics of structures in turbulent reacting flows will aid in understanding the interaction of chemical reactions with the turbulent motion. A better understanding of this key interaction is important for testing existing models of turbulent combustion as well as for suggesting new models.

RESEARCH STATUS

During the past year, progress has been made in several areas of our work on two- and three-dimensional diagnostics. Some specific achievements include the following:

1. Investigation of Differential Diffusion Effects

One of the aspects of reacting flows that makes their complete characterization so difficult is the large number of species present. Even a simple flame contains fuel, oxidizer, intermediates, combustion products, and relatively inert components. In general, each of these components will possess a unique diffusion coefficient. In most of the work in turbulent combustion, however, the simplifying assumption is made that a single diffusion coefficient can be used to characterize all of the species present and that differential diffusion effects can be neglected at reasonably high Reynolds numbers.

A Rayleigh scattering experiment was performed that allowed some of the simplifying assumptions to be checked by providing a direct measurement of differential diffusion effects. In the experiment, hydrogen (which has a Rayleigh cross section lower than that of air) and Freon (which has a higher Rayleigh cross section) were mixed in a ratio so that the effective Rayleigh cross section of the mixture is precisely the same as that of air. The mixture exited into a slowly coflowing stream of air through a cylindrical nozzle, and the resulting turbulent jet was investigated using planar Rayleigh scattering. Because the Rayleigh cross section of the mixture was the same as that of the air, the Rayleigh signal is independent of the mixing of the H_2/Freon jet with the air. However, if the ratio of H_2 to Freon changes due to the different diffusivities of the components, the Rayleigh signal will vary, with Freon rich regions providing more Rayleigh scattering and H_2 rich regions scattering less than the air or regions of the correct ratio.

The experiment showed that the effect is readily observable and that the magnitude of the effect is larger than that predicted by previous modeling efforts. Measurable differential diffusion is found at Reynolds numbers as high as 20,000 and as far downstream as 30 nozzle diameters. In addition to H_2/Freon/air experiments, differential diffusion is also observed when a mixture of H_2 and CH_4 is issued from the nozzle. (H_2 and CH_4 have less disparate diffusion coefficients than H_2 and Freon and may have more implications for combustion work.)

Although the experiment to measure these effects was proposed nearly ten years ago, previous attempts to make the measurement using single-point techniques were not successful. Two critical aspects in the success of the current experiment were (1) the high sensitivity imaging capability of the CCD detector and (2) the use of a second CCD detector to ensure that the Rayleigh cross section of the H_2/Freon mixture was precisely the same as that of air. The results of the experiments have implications for both theorists and experimentalists. A manuscript describing these experiments is in preparation.

2. Development of a New Technique for Visualizing Supersonic Flows.

In a series of experiments performed in collaboration with Robert Dibble of Sandia Laboratories and Godfrey Mungal of Stanford, a simple Rayleigh-scattering technique was developed for visualizing mixing and shock structure in supersonic flows. The elastically scattered light from a thin laser sheet intersecting an underexpanded air-into-air jet was imaged onto a CCD detector. A small amount of humidity in both the nozzle and ambient air resulted in the formation of very small condensed-phase water droplets. The resulting digital images clearly showed the Mach disk and triple-point shock structures as well as large-scale structures near the edge of the jet and along the slip discontinuity. The ability to visualize regions of rapid mixing in this simple way should be a valuable tool for studying supersonic mixing.

The relationship of the signal intensity to the flow properties is complex since the temperature and pressure of the gas vary as do the number density and size distribution of the particles. However, the large dynamic range provided by the CCD detector allows some information on the nature of the condensed-phase particles to be obtained. An investigation of the pixel-to-pixel signal and noise characteristics within a small area of the flow containing particles (in which the particle size and number density can be expected to be constant) revealed that (1) the signal is larger than in regions not containing particles by as much as a factor of 100 and (2) the signal/noise ratio is much smaller than in regions containing no particles. We have used this information in several ways to estimate the characteristics of the particles formed. First, the number of condensed-phase scatterers was determined by considering the noise statistics of particle-containing regions. Second, the magnitude of the signal was used along with the expression for the Rayleigh scattering cross section as a function of particle radius to provide an estimate of the size of the scattering particles. We estimate that the largest of the particles have a radius of 30 nm and, therefore, have very little slip relative to the gas flow, even at supersonic velocities. Finally, by considering the total number of water molecules present in

the flow before condensation, a check on the validity of the results of particle size and number density is obtained as well as some insight into the condensation process responsible for forming particles.

3. Time Evolution of Turbulent Premixed Flames

One of the goals of our research is to measure the temporal evolution of large-scale structures in turbulent reacting flows. One of the main difficulties in realizing this goal is the high speed operation required of both the laser source and the detector. In our previous AFOSR work, we were able to use a high speed framing camera and a CW argon-ion laser to record the Lorenz-Mie scattering from an aerosol seeded premixed flame at an image repetition rate of 48 kHz. The series of images provides useful information on convection velocities and burning velocities. The main drawbacks of this approach, however, are the dependence on marker particles to infer the behavior of the flames and the relatively limited image quality available from the framing camera.

In a current experiment, we are trying to obtain the same information without the limitations imposed by our previous experimental configuration. A double pulsed Nd:YAG laser (pulse separation of 100 ms) is used to illuminate the flow, and Rayleigh scattered light from molecules in the flow is collected by the imaging optics. A high speed rotating mirror displaces subsequent images onto different portions of an intensified CCD detector. Although only a single pair of images is produced, burning and convection velocities can still be obtained, and the quality of the data is considerably improved.

4. Development of a Technique for Measuring the Three-Dimensional Time Evolution of Turbulent Flames

As turbulence is inherently three dimensional, ambiguities remain with measurements in only two dimensions, and quantities such as the scalar gradient and flame curvature cannot be completely determined. The lack of three-dimensional information also leads to uncertainties in

the velocities determined in our time sequence data. We are, therefore, developing a technique for recording a pair of three-dimensional data sets separated in time by a short (100 μs) interval.

The technique is based on the use of a series of closely spaced parallel laser illumination sheets of different wavelengths. A single CCD detector is used with color filters and image displacing wedges which cause the elastic scattering from each illumination sheet to be imaged onto a separate region of the detector. To obtain temporal information, a high speed rotating mirror is again used to displace the images onto different regions of the detector.

In our initial experiment, four laser sheets are formed from the second-harmonic output of a Nd:YAG laser and from the first-, second-, and third-order stimulated Raman scattering from a liquid filled cell pumped by a portion of the 532 nm beam. The laser is double pulsed, and the Lorenz-Mie scattering from an aerosol-seeded premixed flame is recorded at two times separated by an interval of 100 μs.(i.e., a total of eight images is recorded on the CCD detector). This data should allow unambiguous determination of the burning velocity in premixed flames as well as investigation of the effect of curvature on the burning velocity.

PUBLICATIONS

1. M.B. Long, K. Lyons, and J.K. Lam, "Acquisition and Representation of Two- and Three-Dimensional Data from Turbulent Flows and Flames," Computer 22 (8), 39 (1989).

2. M. Winter and M.B. Long, "Two-Dimensional Measurements of the Time Development of a Turbulent Premixed Flame," Combust. Sci. Tech. 66, 181 (1989).

3. A.R. Kerstein, R.W. Dibble, M.B. Long, B.Yip, and K. Lyons, "Measurement and Computation of Differential Molecular Diffusion in a Turbulent Jet," Proceedings of the Turbulent Shear Flows Conference, Stanford, CA, August 1989.

PROFESSIONAL PERSONNEL

Kevin Lyons, Graduate Student
Sumit Sen, Graduate Student
Ruichen Jin, Research Associate

DEGREES AWARDED

None

PATENTS

None

COUPLING ACTIVITIES

Some of our results were presented at the following conferences in 1989:

HTGL Seminar, Stanford, CA, March 15.

13th Meeting of the Sandia Cooperative Group on Thermochemistry of Turbulent Flames, University of California at San Diego, La Jolla, CA, March 16-17.

Gordon Research Conference, Plymouth, NH, July 17-21.

Combustion Dynamics Workshop/Review, Lawrence Berkeley Laboratory, Berkeley, CA, October 16-17.

8th International Congress on Applications of Lasers and Electro-Optics (ICALEO), Orlando, FL, October 16-18.

14th Star Sandia Turbulence and Aerothermochemistry Research Meeting, Nashville, TN, November 6-7.